

# Entanglement of systems of dipolar coupled nuclear spins at the adiabatic demagnetization

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**Abstract.** We consider the adiabatic demagnetization in the rotating reference frame (ADRF) of a system of dipolar coupled nuclear spins  $s = 1/2$  in the external magnetic field. The demagnetization starts with the offset of the external magnetic field (in frequency units) from the Larmor frequency being several times greater than the local dipolar field. For different subsystem sizes, we have found from numerical simulations the temperatures at which subsystems of a one-dimensional nine-spin chain and a plane nine-spin cluster become entangled. These temperatures are of the order of microkelvins and are almost independent of the subsystem size. There is a weak dependence of the temperature on the space dimension of the system.

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## 1. Introduction

Entangled states are very important for quantum computing, teleportation, cryptography [1]. Although entanglement is a profound concept of quantum information theory its experimental realization in many-body systems is an unsolved problem up to now. Meanwhile the existing criterions of the existence of entanglement [2, 3] allow us to investigate entangled states in conventional NMR experiments [4]. It is well known that entanglement emerges in systems of nuclear spins at microkelvin temperatures [5, 6]. Such temperatures can be achieved with the adiabatic demagnetization in the rotating reference frame (ADRF) [7].

In the present article we consider the emergence of entanglement in the course of the ADRF in the system of nuclear spins coupled by the dipole-dipole interaction (DDI). We perform numerical calculations for a chain consisting of nine spins and a plane nine-spin cluster in order to prove the emergence of entanglement of its different subsystems. We use the Wootters criterion [2] for the investigation of the spin pair entanglement. Entanglement of subsystems with larger numbers of spins is investigated with the positive partition transposition (PPT) criterion [3]. Entanglement emerges at approximately the same temperature of the order of microkelvins for all subsystem sizes. However, the temperature depends on the space dimension of the system.

## 2. The density matrix of a spin system at the ADRF

We consider a system of  $N$  nuclear spins  $s = 1/2$  coupled with the external magnetic field. The system is irradiated by the frequency-modulated radio-frequency (rf) field,  $w(t)$ , which is perpendicular to the permanent magnetic field. The Hamiltonian,  $H_{lab}$ , of the system in the laboratory frame can be written as follows

$$H_{lab} = w_0 I_z + H_{dz} + 2w_1 I_x \cos \left[ \int_0^t w(t') dt' \right], \quad (1)$$

where  $w_0$  is the Larmor frequency,  $w_1$  is the amplitude of the rf field (in frequency units),  $I_{n\alpha}$  is the projection of the angular momentum operator of the  $n$ -th spin ( $n = 1, 2, \dots, N$ ) on the  $\alpha$  axis ( $\alpha = x, y, z$ ),  $I_\alpha = \sum_{n=1}^N I_{n\alpha}$ , and  $H_{dz}$  is the secular part of the DDI Hamiltonian [4] which can be written as

$$H_{dz} = \sum_{i < j} D_{ij} (3I_{iz} I_{jz} - \vec{I}_i \vec{I}_j), \quad (2)$$

where  $D_{ij}$  is the DDI coupling constant of spins  $i$  and  $j$ , and  $\vec{I}_i \vec{I}_j = I_{ix} I_{jx} + I_{iy} I_{jy} + I_{iz} I_{jz}$ . The spin dynamics of the system is determined by the density matrix,  $\rho(t)$ , whose time evolution occurs according to the Liouville equation ( $\hbar = 1$ ) [4]

$$i \frac{d\rho(t)}{dt} = [H_{lab}, \rho(t)]. \quad (3)$$

Converting the density matrix,  $\rho(t)$ , with the unitary transformation

$$\rho(t) = e^{-iI_z \int_0^t w(t') dt'} \rho^*(t) e^{iI_z \int_0^t w(t') dt'} \quad (4)$$

and neglecting the irrelevant terms oscillating with the double Larmor frequency one obtains the evolution equation for the density matrix  $\rho^*(t)$

$$i\frac{d\rho^*(t)}{dt} = [(w_0 - w(t))I_z + H_{dz} + w_1I_x, \rho^*(t)]. \quad (5)$$

According to (5) the Hamiltonian,  $H$ , of the system can be written in the rotating reference frame (RRF) as

$$H = \Delta(t)I_z + H_{dz} + w_1I_x, \quad (6)$$

where  $\Delta(t) = w_0 - w(t)$  is the resonance offset of the longitudinal frequency from the Larmor frequency. In the course of the ADRF, the offset,  $\Delta(t)$ , changes slowly in order to satisfy to the adiabatic condition [4]

$$\frac{|\dot{\Delta}(t)|}{\pi w_1^2} \ll 1. \quad (7)$$

The condition (7) means that the resonance offset,  $\Delta(t)$ , changes so slowly that the spin system is in the quasi-equilibrium state at every moment of time [4]. Thus the spin system can be described by the thermodynamic quasi-equilibrium density matrix,  $\rho_{eq}(t)$ , as

$$\rho_{eq}(t) = e^{-\beta H} / Z, \quad (8)$$

where  $\beta$  is proportional to the inverse temperature ( $\beta = \hbar/kT$ ) and  $Z$  is the partition function. It is important to emphasize that the temperature,  $T$ , is the spin temperature of the system which is isolated from all other degrees of freedom. The reason of such isolation is long spin-lattice relaxation times which exceed spin-spin relaxation times by several orders of magnitude [4]. The important consequence for our approach is the following. The decoherence effects at entanglement generation with ADRF are irrelevant.

The entropy,  $S$ , of the system is given by [4]

$$S = -k \operatorname{tr} \{ \rho_{eq}(t) \ln [\rho_{eq}(t)] \}. \quad (9)$$

We start the ADRF with  $\Delta \gg w_{loc}$  where  $w_{loc} = \{ \operatorname{tr} \{ H_{dz}^2 \} / \operatorname{tr} [I_z^2] \}^{1/2}$  is the local dipolar field. Then the offset,  $\Delta(t)$ , is the linear function of time

$$\Delta = \Delta_0 - at \quad (10)$$

where  $\Delta_0$  and  $a$  are the given quantities. Since the entropy,  $S = \text{const}$ , one can obtain the inverse temperature,  $\beta(t)$ , in the course of the ADRF, if the offset,  $\Delta(t)$ , is known. It means that we obtain the density matrix,  $\rho_{eq}(t)$ , during the ADRF. In particular, the entropy,  $S$ , is

$$S = kN \ln 2 + kN \ln [\cosh \left( \frac{\beta \Delta}{2} \right)] - \frac{k}{2} N \beta \Delta \tanh \left( \frac{\beta \Delta}{2} \right) \quad (11)$$

at  $\Delta \gg w_{loc}$ . (11) can be used in order to find the initial inverse temperature of the system.

### 3. The reduced density matrix of a spin pair at the ADRF and the Wootters criterion

In order to obtain the reduced density matrix of an arbitrary pair of spins  $i$  and  $j$  we use the approach developed in [5, 6]. The density matrix,  $\rho_{eq}(t)$ , of (8) can be represented as [5, 6]

$$\rho = \sum_{\xi_1, \xi_2, \dots, \xi_N=0}^3 \alpha_{12\dots N}^{\xi_1 \xi_2 \dots \xi_N} x_1^{\xi_1} \otimes \dots \otimes x_N^{\xi_N}, \quad (12)$$

where  $N$  is a number of spins in the system,  $\xi_k$  ( $k = 1, 2, \dots, N$ ) is one of the values  $\{0, 1, 2, 3\}$ ,  $x_k^0 = I_k$  is the unit matrix of the dimension  $2 \times 2$ ,  $x_k^1 = I_{kx}$ ,  $x_k^2 = I_{ky}$ ,  $x_k^3 = I_{kz}$ , and  $\alpha_{12\dots N}^{\xi_1 \xi_2 \dots \xi_N}$  is a numerical coefficient. Averaging the density matrix of (12) over all spins except spins  $i$  and  $j$  and taking into account that  $\text{tr}\{x_k^{\xi_k}\} = 0$  ( $k = 1, 2, \dots, N$ ;  $\xi_k = 1, 2, 3$ ) we arrive at the following expression for the reduced density matrix,  $\rho_{eq}^{(ij)}(t)$ , of the  $i$ th and  $j$ th spins

$$\rho_{eq}^{(ij)}(t) = \sum_{\xi_i, \xi_j=0}^3 \alpha_{ij}^{\xi_i \xi_j} x_i^{\xi_i} \otimes x_j^{\xi_j}, \quad (13)$$

where

$$\alpha_{ij}^{\xi_i \xi_j} = \frac{2^{N-2} \text{tr}\{\rho x_i^{\xi_i} x_j^{\xi_j}\}}{\text{tr}\{(x_i^{\xi_i})^2 (x_j^{\xi_j})^2\}}. \quad (14)$$

The coefficients,  $\alpha_{ij}^{\xi_i \xi_j}$ , of (14) can be calculated numerically. Then the reduced density matrix of the pair of spins  $i$  and  $j$  is determined completely. In order to apply the Wootters criterion [2] one should find the "spin-flipped" density matrix,  $\tilde{\rho}_{eq}^{(ij)}(t)$ , which is

$$\tilde{\rho}_{eq}^{(ij)}(t) = (\sigma_y \otimes \sigma_y) [\rho_{eq}^{(ij)}(t)]^* (\sigma_y \otimes \sigma_y) \quad (15)$$

where the asterisk denotes complex conjugation in the standard basis  $\{|00\rangle, |01\rangle, |10\rangle, |11\rangle\}$

and the Pauli matrix  $\sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$ . The calculation of the density matrix,  $\tilde{\rho}_{eq}^{(ij)}(t)$ ,

and the diagonalization of the matrix product  $\rho_{eq}^{(ij)}(t) \tilde{\rho}_{eq}^{(ij)}(t)$  are performed numerically. The concurrence of the two-spin system with the density matrix  $\rho_{eq}^{(ij)}(t)$  is equal to [2]

$$C = \max\{0, 2\lambda - \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4\}, \quad \lambda = \max\{\lambda_1, \lambda_2, \lambda_3, \lambda_4\} \quad (16)$$

where  $\lambda_1, \lambda_2, \lambda_3$ , and  $\lambda_4$  are the square roots of the eigenvalues of the product  $\rho_{eq}^{(ij)}(t) \tilde{\rho}_{eq}^{(ij)}(t)$ .

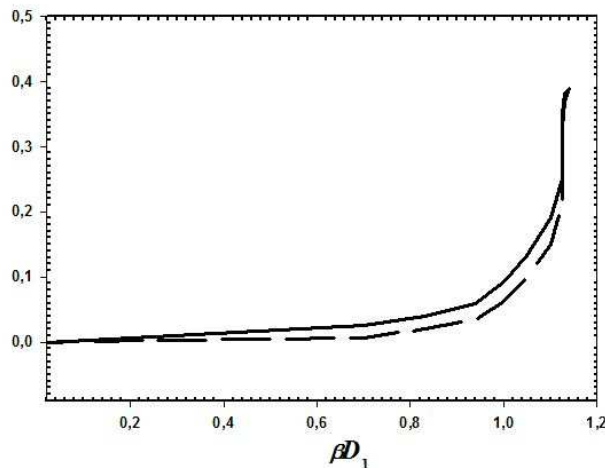
### 4. The entangled state of a spin subsystem with its environment at the ADRF

The spin pair entangled states are a simple type of entanglement which can be described with the Wootters criterion [2] completely. Meanwhile entanglement of bigger

subsystems of the system can also emerge in the course of the ADRF. A necessary condition of separability is described by the Peres criterion [3] which is known as the positive partition transposition (PPT) criterion [3]. According to the criterion, entanglement emerges, if after the transposition of the density matrix over variables of one subsystem there are some negative eigenvalues of the density matrix of the system. We wrote a program which allows us to investigate entanglement of an arbitrary subsystem and its environment in the nine-spin chain and in the square nine-spin cluster in the course of the ADRF on the basis of the criterion [3]. The PPT predicts the emergence of the spin pair entanglement at the same temperature as the Wootters criterion [2]. The PPT criterion uses the sum of the absolute values of negative eigenvalues after the partial transposition of the density matrix as the measure of entanglement. This measure is called the double negativity [8]. We use the criterions of entanglement [2, 3] for the investigation of the entangled states in a nine-spin chain and a plane nine-spin cluster in the next section. Our choice of nine-spin systems is limited by available computational power. Notice that even best available supercomputers do not allow one to calculate spin dynamics when the number of spins in the system exceeds 16 [9]. It is worth to noticing that we consider spin-spin interactions of all spins in contrast to the approximation of the nearest neighbor interactions when the number of spins in the system can be significantly increased [10].

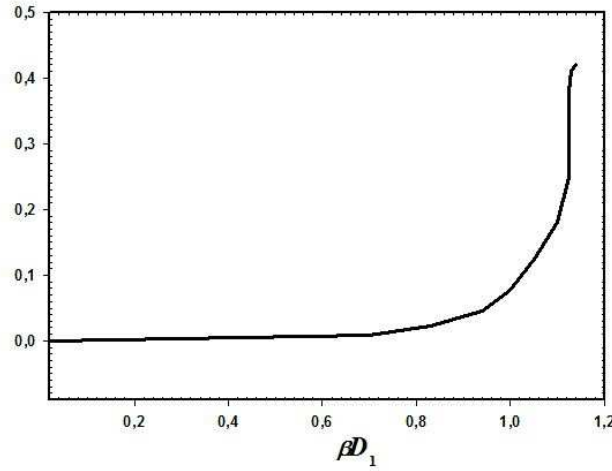
## 5. Numerical analysis of entanglement in a nine-spin chain at the ADRF

The results of the numerical investigation of the spin pair entanglement for a linear chain consisting of nine spins coupled by the DDI at the ADRF are represented in figure 1. We started the ADRF with the offset  $\Delta_0 = 10D_1$  where  $D_1$  is the DDI



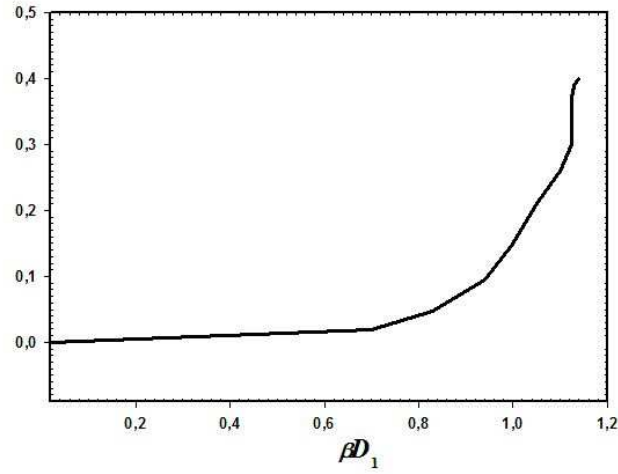
**Figure 1.** The concurrence,  $C$ , versus the dimensionless inverse temperature,  $\beta D_1$ , for spins 1 and 2 of the nine-spin chain according to the Wootters criterion. The dash line shows the same result obtained with the PPT criterion;  $w_1/D_1 = 2$ .

coupling constant of nearest neighbors in the chain. The numerical calculations are performed for  $\Delta_n = (10-n)D_1$ ,  $n = 0, 1, \dots, 10$ . The corresponding values of the inverse temperatures,  $\beta_n = \hbar/kT_n$  ( $n = 0, 1, \dots, 10$ ), are found from (9) for the dimensionless entropy  $S/k = 0.5$ . Figure 1 shows the concurrence of spins 1 and 2 of the nine-spin chain as a function of the dimensionless parameter  $\beta D_1$ . At comparatively high temperatures the concurrence is equal to zero (see figure 1) and the spin system is in a separable state. When the temperature is getting sufficiently low in the course of the ADRF the concurrence is sharply increasing and entanglement emerges. The entangled state appears at  $\beta D_1 \approx 1.1$  (see figure 1), i. e. at  $T \approx 0.5\mu K$  when  $D_1 = 2\pi 10^4 s^{-1}$ . Notice that ordered states of nuclear spins were observed in a  $CaF_2$  single crystal at microkelvin temperatures [7].

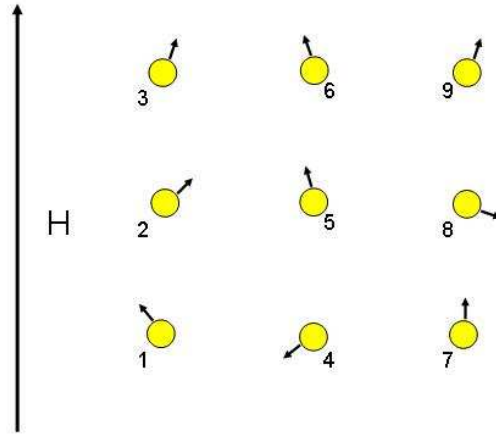


**Figure 2.** The double negativity versus the dimensionless inverse temperature,  $\beta D_1$ , for spin 1 of the nine-spin chain and the other spins;  $w_1/D_1 = 2$ .

The dash line in figure 1 demonstrates that the result for the spin pair entanglement obtained with the PPT criterion coincide practically with the ones obtained with the Wootters criterion. Figure 2 shows the double negativity versus the dimensionless parameter  $\beta D_1$  for the first spin of the chain (the first subsystem) and the other spins of the chain (the second subsystem) in the course of the ADRF when the dimensionless entropy  $S/k = 0.5$ . One can conclude that entanglement emerges at  $\beta D_1 \approx 1.1$ . This result is close to the one for the spin pair entanglement. Figure 3 shows the double negativity versus the parameter  $\beta D_1$  for the first three spins of the chain (the first subsystem) and the other six spins of the chain (the second subsystem) at the same conditions. Here entanglement is getting sufficiently large at  $\beta D_1 \approx 1.1$ . In fact we have found that entanglement of different subsystem emerges approximately at the same temperature.



**Figure 3.** The double negativity versus the dimensionless inverse temperature,  $\beta D_1$ , for the first three spins of the nine-spin chain and the other spins;  $w_1/D_1 = 2$ .



**Figure 4.** The square cluster of nine spins.

## 6. Entanglement in the square cluster of nine spins

The suggested method allows us to study entanglement in the systems of arbitrary space dimensions. As an example, we consider the square cluster consisting of nine spins (see figure 4). The dipolar coupling constant of spins  $j$  and  $k$  (the numbers of spins are

pointed in figure 4) is

$$D_{jk} = \frac{\gamma^2 \hbar}{r_{jk}^3} (1 - 3 \cos^2 \theta_{jk}), \quad (17)$$

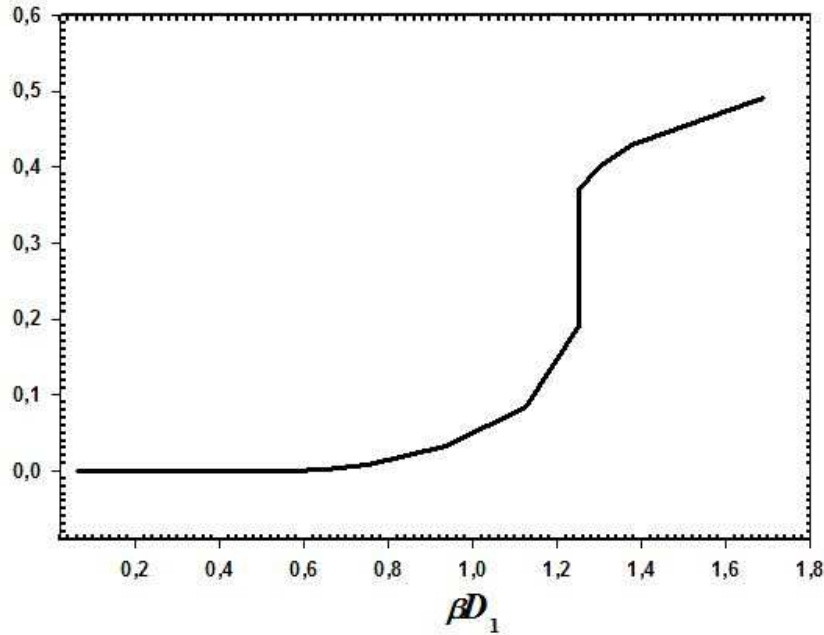
where  $\gamma$  is gyromagnetic ratio,  $r_{jk}$  is the distance between spins  $j$ ,  $k$  and  $\theta_{jk}$  is the angle between the vector,  $\vec{r}_{jk}$ , and the external magnetic field,  $\vec{H}_0$ . The simple analysis yields

$$\cos^2 \theta_{jk} = \frac{9(\{(j-1)/3\} - \{(k-1)/3\})^2}{([\{(j-1)/3\} - \{(k-1)/3\}]^2 + 9(\{(j-1)/3\} - \{(k-1)/3\})^2}, \quad (18)$$

and

$$r_{jk} = a \sqrt{([\{(j-1)/3\} - \{(k-1)/3\}]^2 + 9(\{(j-1)/3\} - \{(k-1)/3\})^2}, \quad (19)$$

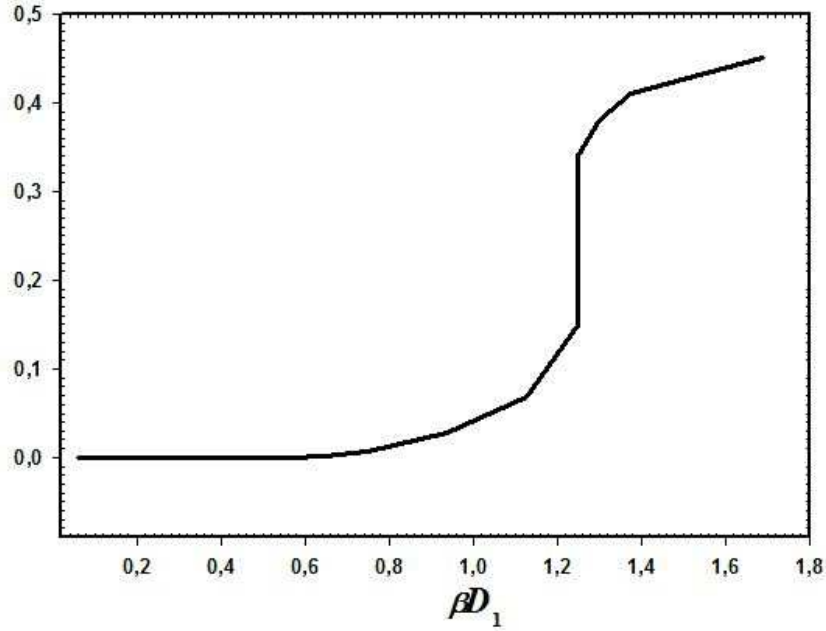
where  $a$  is the distance between the nearest neighbors in the cluster,  $[q]$  is the integer part of  $q$  and  $\{q\}$  is the fractional part of  $q$ .



**Figure 5.** The double negativity versus the dimensionless inverse temperature,  $\beta D_1$ , for spins 1, 2, 4 (first subsystem) and spins 5, 6, 8, 9 (second subsystem), in the square cluster;  $w_1/a = 2$ .

Figure 5 and figure 6 show that the entangled states emerge at  $\beta D_1 = 1.3$  in the two-dimensional cluster. Although the temperatures of the appearance of entanglement are below here than in the one-dimensional case, they are almost the same for different subsystems. The processes of destructive interference are more effective in the two-dimensional cluster than in the one-dimensional chain. They lead to a loss of many-spin correlations which are responsible for the appearance of the entangled states. Thus the temperature of the emergence of entanglement is lower in the two-dimensional cluster than in the one-dimensional chain.





**Figure 6.** The double negativity versus the dimensionless inverse temperature,  $\beta D_1$ , for spins 1, 2 (first subsystem) and spins 3, 6, 9 (second subsystem), at  $w_1/a = 2$  in the square cluster of nine spins.

## 7. Conclusion

We investigated numerically entanglement in the chain of nuclear spins and in the square cluster with the DDI in the course of the ADRF using a special computer program. We showed that the entangled states emerge at microkelvin temperatures for typical DDI coupling constants. Two criterions [2, 3] of entanglement yield the same results for the spin pair entangled states. Entanglement of different subsystems emerges approximately at the same temperature and the pairwise entanglement can be used as an indicator of entanglement of bigger subsystems. It is also worth to notice that we take into account the DDI of the remote spins in contrast to the works [5, 6] where the nearest-neighbor interactions were only considered. The performed calculations show that there are no the entangled states of remote spins both in one-dimensional and two-dimensional cases. Entanglement emerges only if subsystems are in a direct contact. Entanglement of different subsystems at microkelvin temperatures suggest possible applications of linear spin chains in quantum information processing.

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